

REMARKS

Applicants respectfully request reconsideration of the rejection of claims 1-20 as obvious under 35 U.S.C. §103(a) over U.S. Patent No. 5,077,422 to Hara et al.

Claim 1 is directed to a homogeneous process for the hydrogenation of dicarboxylic acids and/or anhydrides. The hydrogenation reaction is carried out in the presence of a catalyst comprising (a) ruthenium, rhodium, iron, osmium or palladium and (b) an organic phosphine. The hydrogenation is carried out in the presence of at least about 1% by weight water, at a pressure of from about 500 psig to about 2000 psig and at a temperature of from about 200 °C to about 300 °C such that from about 1 mol to about 10 mol of hydrogen are used to strip 1 mole of product from the reactor.

It has been found that in the operation of prior art systems for hydrogenation of dicarboxylic acids and/or anhydrides that a decarbonylation reaction occurs in which carbon monoxide is produced from product alcohols or intermediate aldehydes. The carbon monoxide results in deactivation of the catalyst unless the carbon monoxide is removed from the reactor and introduced into a methanation unit for further reaction. In the process of the present invention as claimed, water is available to react with by-product carbon monoxide to produce carbon dioxide and hydrogen via the water-gas shift reaction. The claimed minimum 1% by weight water content can be achieved by adding extra water (e.g., as an additive in the solvent) or it can be achieved by minimizing the quantity of hydrogen used in stripping so that the water generated *in situ* as a byproduct of the hydrogenation reaction is maintained in the liquid phase. Catalyst poisoning caused by

the presence of carbon monoxide is thereby effectively reduced, thus extending the life of the catalyst and improving the economies of the reaction. The carbon dioxide and hydrogen may further react to produce methane, thus eliminating the need for a separate methanation unit. See the application as published, U.S. Pub. No. 20070142679; Paras. [0041] to [0044]. As a further advantage over the prior art, carrying out the hydrogenation reaction in the presence of water eliminates the need to remove water from the starting materials prior to the start of the reaction and allows water to be used as the reaction solvent. See U.S. Pub. No. 20070142679; Para. [0034].

A still further advantage of the process defined by the claims is that the reaction conditions recited therein (i.e., conducting the hydrogenation reaction at a pressure of from about 500 psig to about 2000 psig and a temperature of from about 200 °C to about 300 °C) allow lower gas recycle rates to be used, for instance as little as 1 mol of hydrogen may be used to remove 1 mol of product from the reactor. See U.S. Pub. No. 20070142679; Para. [0037].

Hara et al., referenced at paragraph [0009] of the application as published, disclose a process for producing 1,4-butanediol by hydrogenating succinic anhydride, succinic acid or  $\gamma$ -butyrolactone in the presence of a catalyst comprising ruthenium, an organic phosphine and a phosphorous compound. The phosphorous compound is used as a promoter to improve the activity of the catalyst.

Applicants respectfully submit that the Office has not recognized the full extent of differences between the disclosure of Hara et al. and the present invention as claimed and that the Office has failed to establish a *prima facie* case of obviousness

under 35 U.S.C. §103(a).

In the paragraph bridging pages 3 and 4 of the Office action, the Office asserts that the only difference between the process disclosed by Hara et al. and the claimed invention is that Hara et al. fail to explicitly teach that about 1 mol to about 10 mol of hydrogen are used to strip 1 mol of product from the reactor. The Office has failed to recognize that Hara et al. also fail to disclose a process wherein the reaction is carried out in the **presence of at least 1% water**. Hara et al. disclose at col. 4, lines 54-61 that

When succinic anhydride or succinic acid is used as the starting material, water will be formed as a byproduct, as the reaction proceeds. If the reaction is conducted while **removing such byproduct water** out of the reaction system by a reaction mode such as stripping, for example, by continuously circulating hydrogen in the reaction system, **a further improved reaction activity can be attained**. After completion of the reaction, desired 1,4-butanediol can be obtained from the reaction solution by a usual separating means such as distillation or extraction. (emphasis added)

Nowhere do Hara et al. disclose that water should be present in an amount of 1% by weight, nor recognize the potential for reducing catalyst poisoning and other attendant benefits. Rather, the reference explicitly teaches away from the claimed process by disclosing that byproduct water should be continuously **removed** from the reaction system. The skilled artisan understands Hara et al. to teach that the system will operate best under the driest conditions, that is, that the system should be operated with an amount of hydrogen sufficient to strip out all of the water formed during the reaction.

Significantly, in the main example of Hara et al. in which a dicarboxylic acid or anhydride is used as a feed (succinic anhydride in Example 9), the hydrogen to product ratio is approximately 107:1.<sup>1</sup> Thus, Hara et al. teach the use of a vast excess of hydrogen. The large molar excess of stripping hydrogen to product (as compared to the claimed ratio of about 1:1 to about 10:1) indicates to the skilled person that a large amount of hydrogen is used to strip out the byproduct water from the system and reduce the water concentration to as far as possible and preferably to near zero. Such teaching is contrary to the process as claimed which does not require that the water be stripped out but, rather, requires that at least 1% of water by weight be present during the hydrogenation reaction to achieve the benefits attendant the presence of water discussed above.

Further, on page 4 of the Office action, the Office asserts that "given the pressure ranges of hydrogen disclosed by Hara, those of ordinary skill would expect that the claimed molar ratio of hydrogen to product would [sic: be] covered by the disclosed process, and thus, prima facie obvious." The Office

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<sup>1</sup> Calculated assuming a 100 percent conversion of succinic anhydride (Hara et al. quote 99.4% conversion) and starting with 0.15 mols succinic anhydride, of which 0.047 mols were converted to 1,4-butanediol, 0.0961 mols were converted to  $\gamma$ -butyrolactone and 0.0041 mols were converted to tetrahydrofuran, releasing 0.047 mols of water, 0.0961 mols of water and 0.0082 mols of water, respectively, in the process. The amount of hydrogen gas fed through the reactor is determined by the product of the GHSV, the volume of the reactor and the time (i.e.  $250 \times 0.2 \times 3$ ) giving a total of 16.67 mols of hydrogen of which 0.3993 mols were used in the hydrogenation, leaving 16.26 mols in the stripping reaction. This then gives a hydrogen to product ratio of about 107:1.

fails to articulate any reasoning why the pressures disclosed in Hara et al. are result-effective such that the system disclosed therein requires about 1 mol to about 10 mol of hydrogen to strip 1 mole of product from the reactor. To the contrary, as discussed above, Hara et al. teach that the amount of water present during the reaction should be minimized (i.e., that a large excess of hydrogen used as a stripping gas is required as consistent with Example 9 of Hara et al.).

In view of the above, applicants respectfully request allowance of pending claims 1-20.

Applicants authorize the Commissioner to charge any government fees that may be required, including a three-month extension of time to Deposit Account No. 19-1345.

Respectfully submitted,

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